

RESPONSE UNDER 37 C.F.R. § 1.116  
U.S. Appln. No. 09/249,100  
Attorney Docket No. Q53271

### **REMARKS**

The Examiner has not acknowledged Applicants' claim for foreign priority. Applicants respectfully request that the Examiner make such acknowledgement in the Examiner's next communication.

The Examiner has not returned signed copies of Forms PTO/SB/08 A&B filed with Applicants' Information Disclosure Statements of December 10, 1999, and August 2, 2000. Applicants respectfully request that the Examiner initial and date the Forms, and return signed copies to the undersigned in the Examiner's next communication.

Claims 1-76, 79, 82-85, 87 and 88 are all the claims pending in the application.

At page 2 of the Office Action, Claims 76, 79, 81-85 and 87 have been rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Gratzel et al. (US 4,847,231), optionally in view of Bankmann et al. (US 5,387,726) or Umemura et al (US 4,177,161).

Applicants respectfully traverse the rejection for at least the following reasons.

Applicants wish to point out that Claim 81 has been canceled in the Amendment Under 37 C.F.R. § 1.111, previously filed on July 17, 2002.

Applicants wish to emphasize that the content of the OH group is a content of that chemically bonded to TiO<sub>2</sub> used as a carrier, that is, before the ruthenium compound is formed thereon.

None of the cited references disclose or suggest the presently claimed range of the content of the OH group chemically bonded to TiO<sub>2</sub>.

The OH group content in the TiO<sub>2</sub> carrier usually depends on the method of producing titanium oxide. The chemically bonded OH group cannot be formed by simple wetting,

namely, when titanium oxide is wetted with water, the content of the OH group chemically bonded does not change.

In the ordinary method for preparing  $\text{TiO}_2$ , the  $\text{TiO}_2$  contains OH groups in a small amount because anatase  $\text{TiO}_2$  is converted into rutile  $\text{TiO}_2$  at a temperature higher than that at which OH groups are removed from  $\text{TiO}_2$ . Specifically, when titanium oxide is heated to a high temperature, such as  $700^\circ\text{C}$  (Bankmann et al.) or  $900^\circ\text{C}$  or above (Umemura et al.) to convert anatase  $\text{TiO}_2$  into rutile  $\text{TiO}_2$ , the content of the OH group chemically bound begins to decrease due to dissociation of the OH group from  $\text{TiO}_2$  by dehydration as  $\text{H}_2\text{O}$ .

Specifically, Umemura et al. discloses use of rutile titanium dioxide as a support of a catalyst. Further, Umemura et al. discloses that the rutile titanium dioxide is obtained by calcining anatase titanium dioxide containing a vanadium compound at  $650$  to  $1500^\circ\text{C}$ . Particularly, when a vanadium containing compound is not present, the calcination temperature is approximately  $900^\circ\text{C}$  or more (column 3, lines 3 to 34, particularly lines 26-29).

As explained above, at a temperature as high as  $900^\circ\text{C}$ , the OH group is dissociated from titanium oxide. For this reason, Applicants respectfully submit that in Umemura et al., the amount of the OH group chemically bound to the rutile titanium oxide is lower than that in the presently claimed invention. Further, Umemura et al. is silent on the amount of the OH group chemically bound to titanium dioxide.

Applicants also respectfully submit that in Bankmann, the OH group is also dissociated from  $\text{TiO}_2$  at  $700^\circ\text{C}$  for the reason mentioned above.

In contrast,  $\text{TiO}_2$  having a relatively high content of OH groups as in the present claimed invention is prepared by a method different from the ordinary method wherein

calcination at a high temperature is not required. Accordingly,  $\text{TiO}_2$  of the present invention is different from the rutile  $\text{TiO}_2$  of Gratzel.

Applicants respectfully submit again that catalytic activity depends on the amount of OH group bound to Ti of  $\text{TiO}_2$  used as a raw material for preparing the catalyst. Therefore, the difference of the catalytic activity shows that the structures of the catalyst are different from each other.

In an article co-authored by one of the inventors of the '231 Patent (Gratzel), Mat. Res. Soc. Symp. Proc., Vol. 139, pp. 373 (1989) which was submitted in the IDS filed April 5, 1999, data was reported concerning catalyst in which  $\text{RuO}_x$  is supported on  $\text{TiO}_2$ . In connection with that article, particles of  $\text{RuO}_x$  were formed on  $\text{TiO}_2$  of 100% rutile form or of a mixture of the rutile and anatase forms as shown in electronic microscope photographs. Applicants respectfully submit that, because the '231 Patent is very unclear with respect to preparation of the catalyst, it is appropriate to refer to this article.

In contrast, as mentioned in Applicants' previous responses, it is clear that  $\text{RuO}_2$ /rutile  $\text{TiO}_2$  catalyst in accordance with the claimed invention, in which  $\text{RuO}_2$  particles are not observed on a rutile  $\text{TiO}_2$  carrier (as shown in a photograph in the Second Declaration filed August 13, 2000), can be obtained by supporting  $\text{RuO}_2$  on rutile  $\text{TiO}_2$  having a high OH group content.

Therefore, with reference to the above-mentioned article, which Applicants believe is appropriate, it is clear that the catalyst of the claimed invention is different from the catalyst disclosed in Gratzel.

It should also be noted that in the abstract of this article, it is indicated that the metal particles are selectively dispersed on the  $\text{TiO}_2$  crystallites with the rutile structure. Thus, it is clear that the  $\text{RuO}_2$  is dispersed on the rutile  $\text{TiO}_2$ .

This means that the supported  $\text{RuO}_2$  cannot be highly dispersed since the amount of OH groups on the  $\text{TiO}_2$  is too small. This is demonstrated in Examples 8 and 17 in the specification of the present application. As a result, as shown in the article (page 329, Fig. 1b), the image of  $\text{RuO}_2$  particles is observed in the electron microscope photograph.

Namely, this difference in the catalytic activities is caused by the difference of the amount of OH group bound to Ti of the  $\text{TiO}_2$  used as a raw material for preparing the catalyst, even if the amount of OH group contained in the catalyst as a final product is the same as that contained in another catalyst prepared by using  $\text{TiO}_2$  (rutile 100%) having a different amount of OH content in the  $\text{TiO}_2$  used for preparing the catalyst. (It is actually impossible to measure the OH content in the catalyst as the final product.) The structures are different from each other because the catalytic activities are different from each other.

In view of the above, Applicants respectfully submit that the catalyst of the claimed invention is different from the catalyst disclosed in Gratzel.

To further demonstrate the dependency of the amount of the OH group in the  $\text{TiO}_2$  carrier on the catalytic activity of the catalyst (final product), Applicants herewith submit another Declaration pursuant to 37 C.F.R. § 1.132 executed by Mr. Takuo HiBi, a co-inventor of the present invention.

Specifically, in the present Declaration, a catalyst (Experiment) was prepared in a similar manner as in Example 5 of the present specification using a titanium oxide powder

PT-101. The content of the OH group chemically bound to TiO<sub>2</sub> of Experiment was determined to be  $1.69 \times 10^{-4}$  mol/g-carrier.

The catalytic activity of Experiment, i.e., chlorine formation per unit weight of the catalyst, was determined in the same manner as in Example 5 of the present specification, and the results along with those of Examples 5, 14 and 15 of the present specification were depicted in the figure.

The contents of ruthenium contained in the catalysts of Examples 5, 14 and 15 of the present specification were 4.7% by weight, and that of Experiment was 4.8% by weight. Applicants submit that all of the contents are essentially same. Further, the rutile content of titanium oxide used as a carrier in each of these catalysts is 100%. In this regard, Applicants wish to point out that TiO<sub>2</sub> used in the Examples of Gratzel et al. is P25 powder (a mixture of 80% anatase and 20% rutile), which is not included in the presently claimed invention. The contents of the OH group chemically bound to TiO<sub>2</sub> in Examples 5, 14 and 15 were  $8.5 \times 10^{-4}$ ,  $6.0 \times 10^{-4}$  and  $3.7 \times 10^{-4}$ , respectively.

As clearly shown in the figure, Experiment, Examples 5, 14 and 15 each having a different content of the OH group of TiO<sub>2</sub> used as a carrier, have different catalytic activities.

That is, the amount of the OH group contained in the titanium oxide carrier used for the ruthenium catalyst affects the activity of the catalyst on chlorine production. Therefore, the structure of the catalyst varies depending on the amount of the OH group of TiO<sub>2</sub> used as a carrier.

Further, Applicants respectfully submit that the present invention provides unexpected superior results. In addition to the two previously submitted Declarations, it is apparent from the figure in the present Declaration, that Examples 5, 14 and 15 wherein the amounts of the

OH group of  $\text{TiO}_2$  used as a carrier fall within the presently claimed range of  $3.0 \times 10^{-4}$  to  $10.0 \times 10^{-4}$  mol/g have higher activity than Experiment wherein the amounts of the OH group is  $1.69 \times 10^{-4}$  mol/g. This effect cannot be expected in light of the disclosure of Gratzel et al., optionally in view of Bankmann et al or Umemura et al.

In view of the above, Applicants respectfully submit that the present invention is not obvious over the cited reference. Accordingly, the Examiner is respectfully requested to reconsider and withdraw the rejection.

At page 4 of the Office Action, Claims 76, 79, 81-85 and 87 have been rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Buysch et al (US 6,001,768), optionally in view of Bankmann et al (US 5,387,726) or Umemura et al (US 4,177,161).

Applicants respectfully traverse the rejection for at least the following reasons.

Applicants wish to point out that Claim 81 has been canceled in the Amendment Under 37 C.F.R. § 1.111 previously filed on July 17, 2002.

Buysch discloses use of rutile  $\text{TiO}_2$  as the Examiner pointed out. However, Buysch fails to teach or suggest not only the amount of OH group chemically bound to  $\text{TiO}_2$  used as a carrier, but also the dependency of the catalytic activity on the amount of OH group.

In contrast, as shown in the present Declaration, the catalyst having the presently claimed range of the amount of the OH group chemically bound to titanium dioxide has unexpectedly superior activity.

Further, as set forth above, Bankmann et al and Umemura et al do not rectify the deficiencies of Buysch.

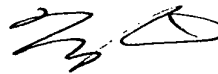
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In view of the above, Applicants respectfully submit that the present invention is not obvious over the cited reference. Accordingly, the Examiner is respectfully requested to reconsider and withdraw the rejection.

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,



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Fang Liu  
Registration No. 51,283

SUGHRUE MION, PLLC  
Telephone: (202) 293-7060  
Facsimile: (202) 293-7860

WASHINGTON OFFICE

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